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## **ENHANCED THERMAL CONDUCTIVITY OF EPOXY MATRIX COMPOSITES FILLED WITH BORON NITRIDE PARTICLES**

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### **ABSTRACT**

The present paper deals with the estimation of thermal conductivity of epoxy Composites embedded with boron nitride (BN) micro-fillers. These composites with BN content ranging from 0 to 11.3 vol. % have been prepared and the thermal conductivities of the samples are measured experimentally. A numerical simulation using finite element package ANSYS is used to explain heat transfer process within epoxy matrix filled with micro-BN and the effective thermal conductivity values obtained from this method are validated with experimental results and theoretical correlation. It is observed that for 11.3 vol% of micro-BN in epoxy matrix, the increase in thermal conductivity is about 27.82 % while for 30 vol% the increase in thermal conductivity is about 440 % which is reasonably higher compared to neat epoxy resin. The results show that the BN particles show a percolation behavior at 20 vol% at which a sudden jump in thermal conductivity is noticed.

### **INTRODUCTION**

It is known that micro-electronic packaging has been playing increasing important role in the rapid progress of the electronic and electrical technologies. With the integration scale of the microelectronic circuit increasing, more and more heat is produced when the circuit works. The heat must be dissipated away in time to avoid over-heat occurrence, which requires that the packaging materials should have good thermal conductivity besides having traditional physic-mechanical properties. Polymers and ceramics have good electrical, mechanical and thermal properties and so there is a growing demand for them as packaging materials [1]. However, common polymers for packaging, such as epoxy, polyester, polyethylene, polypropylene etc. have low thermal conductivities for which they cannot effectively dissipate heat when used in various electronic

devices. Therefore, increasing the thermal conductivity of packaging materials would open up large new markets. It is noted that mostly theoretical models for predicting the effective thermal conductivity of filled polymer composites are based on assumptions for simplicity. For example, Dawson and Briggs [2], Kim and Yoon [3], and Ervin et al. [4] estimated the effective thermal conductivities of composites with uniformly and periodically distributed regular particles. The 2D and 3D finite element models of filled polymer composites developed in previous work [5-7] are also based on the assumptions of randomly distributed spherical particles and/or cylindrical fibers. These models cannot reflect the actual microscopic irregularities of the filler shape and distribution, especially for the case of high filler content when a continuous network of filler is formed. Since the models previously adopted are inadequate for estimating the effective thermal conductivity of filled polymer composites, therefore a theoretical model based on the percolation theory with consideration of the actual shape and distribution of fillers is done in the present research.

Below the percolation threshold, the conductivity is negligible and the threshold conductivity of the composites is equal to the polymer conductivity or slightly higher. However, for thermal conductivity, many authors [8] found that there are significant differences between electrical and thermal conductivity behavior of filled system. Polymers filled with conductive ceramics are thus considered suitable for electronic packaging, encapsulation and for substrates in printed circuit boards. In view of this, the present research is to enhance the thermal conductivity of epoxy composites by filling it with boron nitride (BN) powder. Thermal conductivity of particle filled polymer composites is investigated numerically and experimentally. In the numerical study, the finite-element program ANSYS is used to calculate the thermal conductivity of the composite by using the results of the thermal analysis. Improved thermal conductivity in polymers may be achieved either by molecular orientation or by the addition of conductive fillers Dilek Kumlutas et al (9). It is well known that thermal transport increases significantly in the direction of orientation and decreases slightly in the direction perpendicular to the orientation. Reinforced polymeric materials are being increasingly used in electronic systems due to their ease of manufacturability, light-weight and tailor able properties. As the size of packages becomes smaller they encounter increasingly high temperature Karthik Ramani et al (10). Increased temperature adversely affects the reliability and electrical performance of product. An increase in temperature of approximately 10°C reduce the main time to failure by factor of two. Thermal conductivity of boron nitride-filled polybenzoxazine exhibits a very high conductivity value. It has bimodal particle size distribution which assists in increasing the particle packing density. Hatsuo Ishida et al (11). This filler matrix system provides a highly thermally conductive composite due to the capability of forming conductive networks with low thermal resistance along the conductive paths. The major ingredient in semiconductor molding compounds is filler. These are blended into the polymer matrix to modify and improve select composite properties Procter P. et al (12). Molding compounds formulated to maximum conductivity, using the highest conductivity fillers at the highest feasible volume fraction, did not provide either the expected nor needed thermal performance gain.

Several studies have shown that BN outperforms many other types of ceramic fillers in improving the thermal conductivity of the filled polymers. The purpose of this study was to investigate major factors affecting the thermal conductivity of BN-filled thermoplastic polymers Hsiao Yen Ng et al (13). The effects of filler characteristics, which include particle size, aspect ratio, surface area, and surface modification, on the thermal conductivity of the composites are discussed.

## MATHEMATICAL MODEL

The theoretical analysis of the heat transfer within the particulate filled polymer has been reported by the authors previously [2]. It is based on the following suppositions: (a) the distribution

or dispersion of the solid micro-spheres in the polymer matrix is uniform; (b) the temperature distribution along the direction of heat flow is linear. The expression for effective thermal conductivity of the composite is deduced as:

$$k_{eff} = [1/k_m(1 - \frac{6v_f}{\pi})^{1/3} + 2(k_m(\frac{4\pi}{3v_f})^{1/3} + \pi(\frac{2v_f}{9\pi})^{1/3} \times (k_f \frac{\rho_m}{\rho_f} - k_m))^{-1}]^{-1} \quad (1)$$

Here,  $k_m$ ,  $k_f$  are the respective heat conductivities of the polymer and the filler phase,  $\rho_m, \rho_f$  are densities of the polymer and the filler phase respectively and  $v_f$  is the filler volume fraction.

## EXPERIMENTAL DETAILS

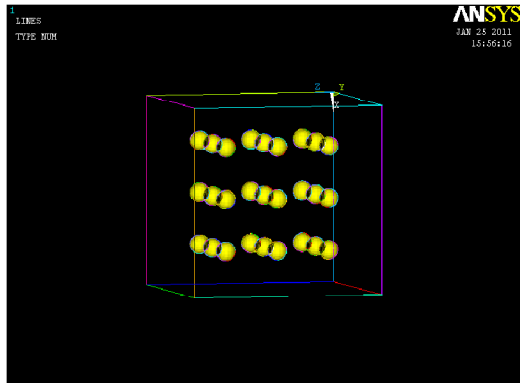
Epoxy is chosen as the matrix primarily of its low density. Boron nitride (100  $\mu\text{m}$ ) is chosen as the filler material. Hexagonal BN having graphite-like crystalline structure is an excellent electrical insulator, because its outer electrons are firmly bound by the nitrogen atoms. In addition, it is a good heat conductor (thermal conductivity 110 W/m-K). The thermal conductivity of BN is significantly higher than that of other ceramic fillers such as fused silica or aluminum oxide. Low temperature curing epoxy LY 556 resin and the hardener (HY951) are mixed in a ratio of 10:1 by weight as recommended. The dough (epoxy filled with BN in different weight proportions) is then slowly decanted into the disc type cylindrical glass molds, coated beforehand with a uniform thin film of silicone-releasing agent. Composites of 12 different compositions (BN content varying from 1.4 to 11.3 vol %) is made. Unitherm™ Model 2022 is used to measure the composite thermal conductivity following ASTM E-1530 standard.

## RESULTS AND DISCUSSION

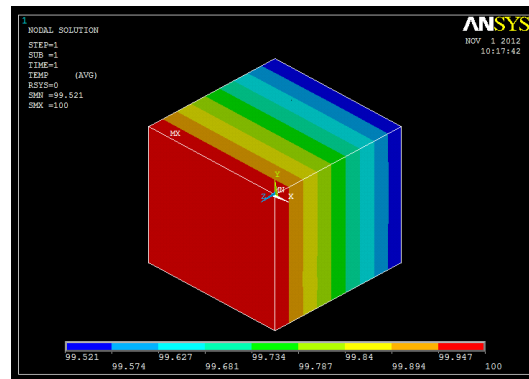
### Numerical Analysis

In the numerical analysis of the heat conduction problems, the temperatures at the nodes along the surfaces ABCD is prescribed as  $T_1$  ( $=1000^\circ\text{C}$ ) and the convective heat transfer coefficient is assumed to be  $2.5 \text{ W/m}^2\text{-K}$  at ambient temperature of  $27^\circ\text{C}$ . The heat flow direction and the boundary conditions are shown in Fig. 4.1. The othersurfaces parallel to the direction of the heat flow are all assumed adiabatic. The temperatures at the nodes in the interior region and on the adiabatic boundaries are unknown. These temperatures are obtained with the help of finite-element program package ANSYS.

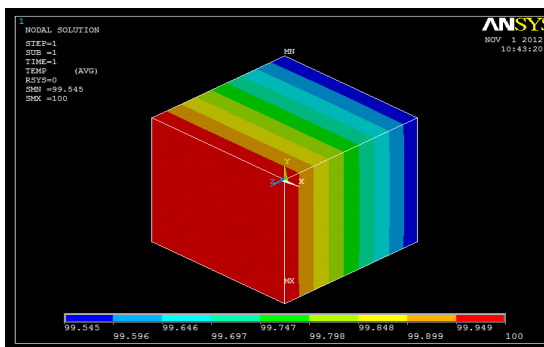
Thermal conductivities of epoxy composites filled with boron nitride particles up to 11.3 % by volume are numerically estimated by using the spheres-in-cube model. A typical 3-D model showing arrangement of spherical fillers with a particle concentration of 1.4 vol% within the cube shaped matrix body is illustrated in Fig.1.1. The temperature profiles obtained from FEM analysis for the composites (spheres-in cube arrangement) with particulate concentrations of 1.4, 3.35, 5.23, 7.85, 9.04 and 11.3 vol. % are presented in Fig. 1.1 - Fig.1.6 respectively.



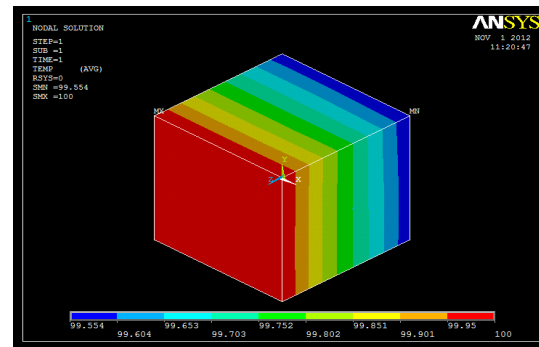
**Fig 1:** Geometric model of boron nitride with particle concentration of 1.4vol%.



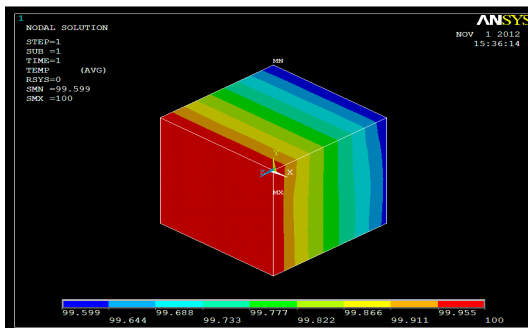
**Fig 1.1:** Temperature profile for composite (spheres) in epoxy matrix (cube) at 1.4vol%.



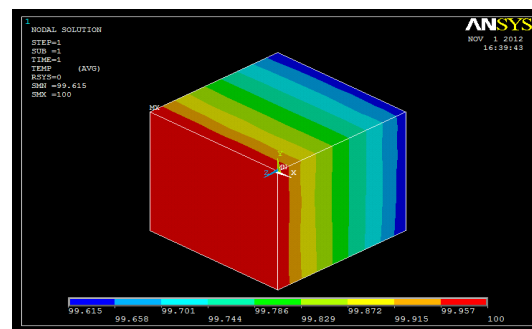
**Fig 1.2:** Temperature profile for composite with particle concentration of 3.35vol%



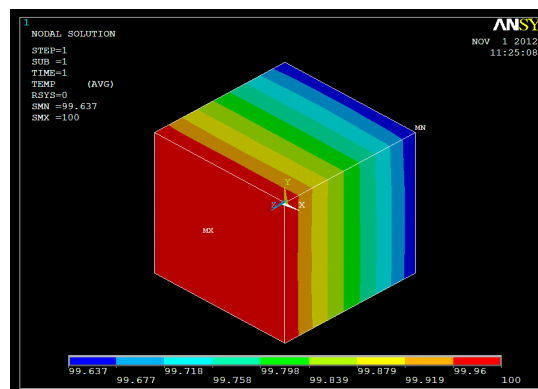
**Fig 1.3:** Temperature profile for composite with particle concentration of 5.23vol%.



**Fig 1.4:** Temperature profile for composite with particle concentration of 7.85vol%.



**Fig 1.5:** Temperature profile for composite with particle concentration of 9.04vol%.



**Fig 1.6:** Temperature profile for composite with particle concentration of 11.3vol%.

The values of effective thermal conductivities of the particulate filled epoxy composites with varied proportions of boron nitride obtained using rule-of-mixture model, Maxwell's equation, Lewis and Nielsen's equation and ROM series model and Geometric model are presented in Table 1. It presents a comparison among the results obtained using these models with regard to the corresponding values of effective conductivity obtained experimentally.

**Table 1:** Effective thermal conductivity values obtained from different methods.

Sample No.	Boron nitride (Vol. %)	Effective thermal conductivity of the composite [W/m.K]				
		ROM series model	Maxwell model	Lewis-Nielsen Model	Geometric Model	Experimental
1	1.4	0.368	0.378	0.385	0.393	0.372
2	3.35	0.376	0.400	0.419	0.439	0.401
3	5.23	0.383	0.422	0.456	0.489	0.407
4	7.85	0.394	0.454	0.514	0.568	0.432
5	9.04	0.398	0.469	0.545	0.608	0.442
6	11.3	0.410	0.499	0.611	0.692	0.464

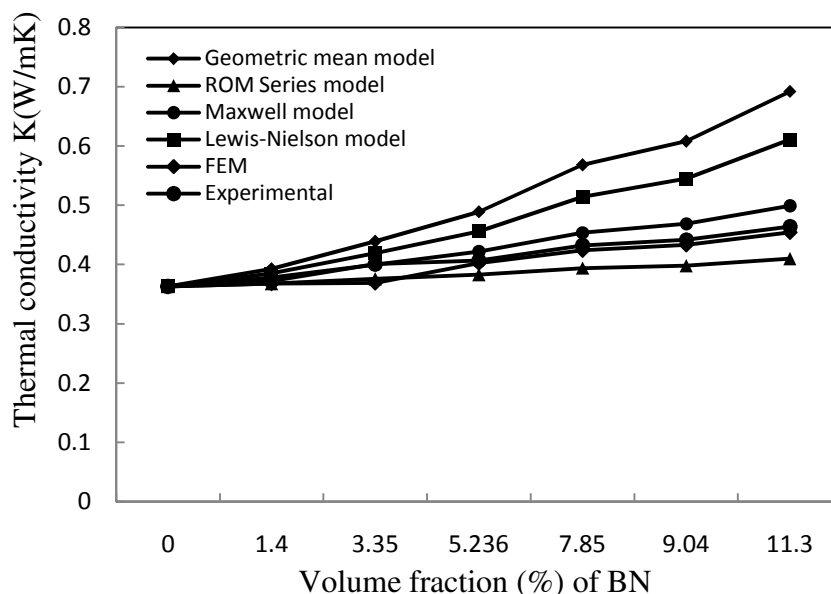
**Table 2:** Percentage error of different models with respect to experimental values

Sample No.	Boron nitride (Vol. %)	Percentage errors with respect to the experimental value (%)				
		ROM series model	Maxwell model	Lewis-Nielsen Model	Geometric Model	Experimental
1	1.4	1.075	-1.613	-3.495	-5.645	0.372
2	3.35	6.234	.2494	-4.489	-9.476	0.401
3	5.23	5.896	-3.685	-12.039	-20.147	0.407
4	7.85	8.796	-5.093	-18.981	-31.48	0.432
5	9.04	9.954	-6.109	-23.30	-37.55	0.442
6	11.3	11.637	-7.543	-31.68	-49.13	0.464

**Table 3** Thermal conductivity values for composites obtained from FEM and Experiment

Sample No.	Boron nitride (Vol. %)	Effective thermal conductivity of composites $K_{eff}$ (W/m-K)		Percentage errors with respect to the experimental value (%)
		FEM (Spheres-in-cube Model )	Experimental	
1	1.4	0.368	0.372	1.075
2	3.35	0.369	0.401	7.980
3	5.23	0.4025	0.407	1.106
4	7.85	0.424	0.432	1.852
5	9.04	0.4335	0.442	1.923
6	11.3	0.4545	0.464	2.047

The simulated values of effective thermal conductivity of the composites obtained by FEM analysis are presented in Table 3 along with the corresponding measured values. The findings for spheres-in-cube arrangements are found to be different as seen in this table.



**Fig. 1.7:** Comparison of thermal conductivity values obtained from different methods.

It is noticed that while the FEM analysis can very well be used for predictive purpose in determining the effective thermal conductivity for a wide range of particle concentrations. The difference between the FEM simulated values and the values obtained by Rule of mixture and Maxwell's model may be attributed to the fact that these models do not consider the distribution pattern of the filler particles within the matrix body, which the FEM analysis does. With addition of 11.3% of 100 micron size boron nitride particle, thermal conductivity of epoxy increased from 0.363W/m-K to 0.464W/m-K. It can be seen from the graph that for less filler concentration, the slope of the curve is less and as the filler volume fraction increases, the curves representing FEM become steeper. It might be due to the fact that with increase in filler concentration, the inter-particle distance reduces and the conductive chains begin to form which increase the thermal conductivity quite reason.

The variation of  $k_{eff}$  with the BN content is shown in Fig.1.7. With increasing BN content in the composite, the value of  $k_{eff}$  keeps increasing. Boron nitride is a solid lubricant that improves the rheological properties of the polymer system. This increases the mobility of the hexagonal BNparticulates which, in turn, promotes their natural tendency to attach themselves to each other by their basal planes. Thermal bridges are formed across these planes throughout the system resulting in a rise in conductivity. The micro-BN filled composite has a much higher thermal conductivity because larger sized filler causes smaller contact resistance and platelet-shaped filler more easily forms conductivity chain. BN filler has a high intrinsic thermal conductivity and show higher thermal conductivity compared to other ceramic filler filled composites at the same filler size and morphology.

## CONCLUSIONS

Incorporation of BN results in enhancement of thermal conductivity of epoxy resin and there by improves its heat conduction capability. With addition of 11.3 vol% of BN (100 micron size), the thermal conductivity improves by about 27.82%. The proposed theoretical model shows good agreement with the experimental results for composites with low filler concentrations.



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